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THE USE OF HIGH-EFFICIENCY GAMMA COUNTING IN CONJUNCTION WITH BETA COUNTING FOR DISCRIMINATION BETWEEN THORIUM AND URANIUM

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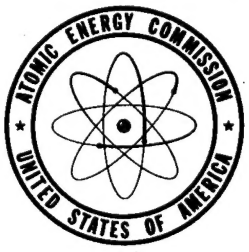

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ABSTRACT

Use of the high-pressure gamma chamber and vibrating reed electrometer to measure gamma activity and the end window Geiger counter to measure beta activity has permitted discrimination between activity due to thorium and that due to uranium so that the thorium and uranium content of ore samples can be determined radiometrically within a factor of 2 and usually within 15 per cent.

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TOPICAL REPORT MITG-219

THE USE OF HIGH-EFFICIENCY GAMMA COUNTING IN CONJUNCTION WITH BETA COUNTING FOR DISCRIMINATION BETWEEN THORIUM AND URANIUM

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I. INTRODUCTION

In order to determine the amounts of both thorium and uranium present in a mixed ore by radioassay methods, at least two measurements must be made and the conditions must be such that the two readings are contributed to in different proportions by the thorium and uranium content.

An unsuccessful attempt was made by this laboratory to discriminate radiometrically between thorium and uranium using a method based upon the self-absorption of beta radiation[1/]. This method had the desirable feature of requiring a minimum of special equipment, but results of useful accuracy were not obtained. There are, of course, other radiometric methods available for distinguishing between the radiations from the thorium and uranium series and, of these, that involving high efficiency gamma counting in conjunction with beta counting seemed to show promise. This gamma-beta method was, therefore, chosen as the subject for further investigation.

If the response of the measuring equipment varies linearly with radiation intensity and the geometry of the beta and gamma measuring systems is standardized, the per cent ThO_2 and per cent U_3O_8 may be obtained from two readings. These readings are substituted in two equations containing four known constants and then these equations are solved simultaneously to yield percentages of ThO_2 and U_3O_8 in the unknown mixture. The four constants, amperes of ionization current per gram of ThO_2 and per gram of U_3O_8 and counts per minute per gram of ThO_2 and per gram of U_3O_8, may be determined from measurements made on one or more samples of ore containing known amounts of U_3O_8 as their only radioactive constituents and one or more samples containing known amounts of ThO_2 and U_3O_8 as their only radioactive constituents.

II. SUMMARY

The gamma-beta method of radiometric discrimination between thorium and uranium was tested on twelve mineral samples. Radioassays for ThO_2 and U_3O_8 were within a factor of 2 of the chemical analyses, and usually within 15 per cent. This method may have some limited usefulness in appraising unknown minerals, and, upon further investigation, might give more accurate results.

1/ Topical Report MITG-208

III. EXPERIMENTAL TECHNIQUES

Measurements of Gamma Activity

The high-pressure ionization chamber, in conjunction with a vibrating reed electrometer, was designed to measure gamma radiation intensity efficiently in the presence of beta radiation. Gamma rays produce relatively little ionization in a gas at atmospheric pressure, so chambers containing compressed gas must be used. The chamber used for the present investigation contained argon at a pressure of approximately 550 psi. The collecting potential was furnished by dry cells delivering a total of about 270 volts which, tests indicated, caused the chamber to operate well within the region of saturation. Samples were placed in a 16-gage steel thimble, extending axially into the chamber and surrounded by the collecting electrode. The relatively thick walls of the thimble minimize the formation of ions by beta radiation, so that the ionization current produced is a practical measure of the gamma radiation alone[2].

In order to measure accurately the small ionization currents produced in the chamber, it is necessary to utilize a sensitive current-measuring device. The vibrating reed electrometer[3] admirably fulfills most of the requirements for an ideal ionization current detector. This device is very simple and dependable in operation, and has extremely low drift and high sensitivity. This instrument is commercially available from the Applied Physics Corporation of Pasadena, California, and the manufacturer claims that currents as small as 10^{-17} ampere, originating in a high-impedance source, may be detected. The combination of the high-pressure gamma chamber, enclosed by lead shielding four inches thick, and vibrating reed electrometer used in our tests gave rise to a background current of the order of 1.6×10^{-13} ampere. This background current, however, proved to be remarkably constant and stable over a period of months and, therefore, it was considered practical to rely on the measurement of currents as low as 0.5×10^{-13} ampere. This would allow 0.032 per cent of U_3O_8 or 0.07 per cent of ThO_2 in a 100-gram sample to be measured.

Thirty grams of each sample was placed in a small beaker which was then lowered into the sample holder of the high-pressure gamma chamber and the ionization current produced by it measured with the vibrating reed electrometer.

The vibrating reed electrometer provides two methods of measuring the current due to the ionization caused by a sample. For currents smaller than about 2×10^{-12} ampere, the rate-of-charge method proves most satisfactory. When ionization is taking place within the chamber, current flows into the electrometer capacitance building up a charge thereon. The voltage across the electrometer capacitance, which is proportional to charge, is indicated by the meter on the control panel of the instrument.

2/ However, extremely active β-emitters can be measured by Bremsstrahlung per report ORNL 314.

3/ The theory of the vibrating reed electrometer is given in the "Design of Dynamic Condenser Electrometer" by Palevsky, Swank and Grenchik, Review of Scientific Instruments, 18, 298-314, May, 1947.

If ΔQ is the change in charge during time Δt, then the average current I_{Av} is

$$I_{Av} = \frac{\Delta Q}{\Delta t} \qquad \text{and, therefore,}$$

$$I_{Av} = \frac{\Delta V}{\Delta t} C .$$

C is the equivalent capacity of the ionization chamber-electrometer system which, for the equipment used in this investigation, was determined to be about 14 micro-microfarads. In making current measurements by the rate-of-charge method, a stop-watch is used to determine the time necessary for the meter on the electrometer control panel to deflect from 10 per cent of full scale to full scale. This time is measured several times and the average used to calculate the current. Because of statistical variations in the current, due to the samples and that due to background, it is not to be expected that the time taken for the meter to deflect 90 per cent of full scale will be constant. Experience gained from making many such measurements, however, has shown that this time, for successive measurements of the same current, does not normally vary more than a few per cent. In conducting these tests, the time for a 90 per cent of full scale meter deflection was measured as many times as necessary to obtain four or five readings which showed a maximum deviation of not more than 5 per cent. Using this procedure, if the current due to a particular sample was measured at various times, the results would rarely disagree by more than 5 per cent. Although this accuracy could probably be improved, at least to a small degree, it was felt that it was adequate for the purposes of this investigation. This is a desirable feature of this method of gamma activity measurement, since it means that the activity of a sample can usually be ascertained in a period of about five minutes.

For very active samples, the steady deflection method is used. This involves the use of a calibrated high value resistor connected between terminals "H" and "G" of the electrometer[4]. When using this resistor, the current originating in the source is given by:

$$I = \frac{E_f}{R} \qquad \text{where:}$$

R = resistance of calibrated resistor

E_f = average deflection of panel meter in volts.

The deflection of the panel meter will, of course, not be steady and its readings must be averaged over a sufficiently long period to assure that an accurate value is obtained. In this investigation, the averaging was done mentally while observing the meter fluctuations for periods of from one to five minutes.

In order to ascertain the effect of sample density or the use of different amounts of the same sample, the currents due to several weights of two

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4/ For this and other details concerning the vibrating reed electrometer see "Instruction Manual for Vibrating Reed Electrometer" furnished by the Applied Physics Corporation.

samples were measured. The results of these measurements indicated that the ionization current per gram of sample was virtually a constant regardless of the amount of sample used, over the range of 2.5 to 250 g. There is good reason to believe, therefore, that the effects of self-absorption or sample geometry are negligible.

Measurements of Beta Activity

The apparatus used to measure the beta activity of the samples analyzed was essentially the same as that described in Topical Report MITG-208. This rack-up apparatus allows sources to be brought within a predetermined distance, which may be precisely duplicated, of the window of an end-window Geiger-Mueller counter. For the present measurements Victoreen counters with thin mica end windows were used and the samples were positioned 2.5 inches from the window. All measurements were made using samples of one gram, placed in cylindrical brass sample holders with an area of 12.5 sq cm. This means that the thickness of the samples used was 0.08 g/sq cm, which is substantially less than infinite thickness for beta radiation. Under these conditions the error introduced by gamma radiation from the sample is virtually negligible. This error would be very small even if samples of infinite thickness were used; but, since it is a simple matter to weigh samples to an accuracy of better than 1 per cent, there is little or nothing to be gained by using samples of infinite thickness.

The counting rates of all but one sample were sufficiently low that resolving time errors were negligible. The euxenite, however, gave a relatively high counting rate and, therefore, it was counted with the aid of a 0.0205-inch aluminum absorber, which reduced the normal counting rate by a factor of 2.74.

All counting rates were obtained from measurements of total counts exceeding 5000 and several such measurements were made on each sample in order to arrive at the average net counting rates given in the tables included herewith. The probable error due to statistical fluctuations in the counting rates, therefore, should be under 1 per cent. The accuracy of counting some of the less active samples might have been increased slightly by using an annular sample holder to surround a thin-wall counter tube but even the most inactive samples gave counting rates sufficiently above background to make this added complication seem unwarranted.

IV. RESULTS OBTAINED BY GAMMA-BETA COUNTING

Deviation of Constants

The constants, T_γ , U_γ , T_β , and U_β , which define the relative gamma and beta activities of thorium and uranium, were determined by beta and gamma measurements of ores whose true chemical composition is believed to be accurately known. For simplicity, the gamma and beta activity of U_3O_8 should be derived from ores known to contain U_3O_8 as their only radioactive component. No such ores were readily available, so two Belgian Congo ore samples known to contain an amount of thorium not exceeding 1.5 per cent of the amount of uranium present were used. Since the amount of thorium present in these ores is known to be small and both the beta and gamma activities of uranium are considerably more than those of thorium, it is evident that the assumption that these ores contain no thorium introduces a negligible error in determining U_γ and U_β . Using U_γ and U_β , derived from these ores, it was possible to determine the beta activities of ThO_2 in an ore containing a high ratio of ThO_2 to U_3O_8. Chemical analyses of the standards used also included analyses of two samples of Canadian pitchblende ores which were used as standards in preliminary tests.

Table 1. Analyzed Standards

	% ThO_2	% U_3O_8
Monazite Sand (National Bureau of Standards Sample)	1.138	0.0448
Belgian Congo Ore G	Trace	1.81
Belgian Congo Ore C	Trace	3.49
NW-194 Eldorado Pitchblende	Trace	1.32
NW-208 Eldorado Pitchblende	Trace	0.806

In the preliminary and final tests different Victoreen end-window counters were used as well as different uranium standards. The values obtained for the constants are given in Table 2 and the deviation is given in the Appendix.

Table 2. Values for Constants

	Preliminary	Final	Units
T_γ	7.00	7.05	x 10^{-13} amp/g ThO_2
U_γ	15.8	14.4	x 10^{-13} amp/g U_3O_8
Ratio U_γ /T_γ	2.26	2.04	
T_β	16,000	12,850	cpm/g ThO_2
U_β	75,200	60,600	cpm/g U_3O_8
Ratio U_β /T_β	4.70	4.72	

The ratios, U_γ / T_γ and U_β / T_β, are such that it should be possible to secure useful discrimination between thorium and uranium. The beta constants with different standard uranium ores have nearly the same ratio and the difference is due to the difference between Geiger tubes used. The change from Eldorado to Congo ores was made because the Eldorado material was suspected of having some radon loss[5]. This affects gamma emission much greater, proportionately, than beta emission and may account for the difference in U_γ for the different sets of standards.

Let I_x = ionization current produced by unknown mixture

N_x = β count produced by unknown mixture

P_u, P_t = fraction of unknown mixture that is U_3O_8, ThO_2

Then:

$$P_t T_\gamma + P_u U_\gamma = I_x$$

$$P_t T_\beta + P_u U_\beta = N_x$$

These two equations may be solved simultaneously to give P_u and P_t.

After deriving the preliminary constants, β and γ measurements were made on several samples prepared from thorium minerals purchased from Wards Natural Science Museum. The minerals were usually diluted with quartz. These samples were used in the previous investigation (MITG-208). The results were good for some samples and poor for others. The accuracy of our chemical analyses was suspected and a new set of samples was made up, including some nonmagnetic fractions of black sands (MITG-201).

The thorium content of the new set of samples was determined by chemical analyses supplied by the U. S. Geological Survey, while the uranium content was determined by analyses made by this Laboratory.

Beta and gamma measurements were made on these samples and the ThO_2 and U_3O_8 contents were calculated with the final values of the constants. Results of the final set are given in Table 3. The final chemical and counter values on the samples in the preliminary set are compared with the preliminary values in Table 4.

5/ "Use of G-M tubes for determination of State of Equilibrium of Radioelements in Uranium Ores". Topical Report No. 2, by C. Lapointe, 7/21/48, Radiation Laboratory Department of Mines & Resources.

Table 3. Chemical vs. Gamma-Beta Activity Results

Ore	Chem. Analyses		Gamma-Beta Activity			
			Thorium		Uranium	
	% ThO_2	% U_3O_8	% ThO_2	% Deviation*	% U_3O_8	% Deviation*
**Aeschynite Blomstrandine (Norway)	6.60	3.86	5.73	-13.2	4.27	+11
**Monazite (Madagascar)	4.29	0.467	3.59	-16.3	0.87	+86
**Polycrase Euxenite (Brazil)	4.65	6.95	3.45	-26	7.82	+12.5
**Euxenite (Norway)	2.20	15.3	2.05	-6.8	17.05	+11
**Thorianite (Ceylon) (1/20) A	3.40	1.30	3.37	-0.9	1.26	-3.1
**Thorianite (Ceylon) (1/15) B	2.36	0.339	1.99	-15.6	0.394	+16
**Thorite Orangite (Madagascar) (1/30)	2.12	0.270	1.63	-23	0.0992	-63
Non Mag Fraction Black Sand A-5	0.80	0.031	0.435	-46	0.0574	+85
Non Mag Fraction Black Sand A-21	0.24	0.051	0.237	-1.25	0.0732	+44
Non Mag Fraction Black Sand A-23	2.68	0.536	3.72	+14	0.705	+31
Non Mag Fraction Black Sand A-25	0.34	0.022	0.220	-35.3	0.0449	+104
Non Mag Fraction Black Sand A-26	1.82	0.315	1.95	+7.1	0.362	+15
Median bias				-14.4		+15.5

Note: Figures in parentheses are ratios of dilution with quartz.

* Deviation of radiometric from chemical.

** From Ward's Natural Science Museum.

Table 4. Comparison of Initial and Final Assays; Chemical and Radiometric

Ore	Chemical				Radiometric			
	Initial		Final		Initial		Final	
	% ThO_2	% U_3O_8	% ThO_2*	% U_3O_8	% ThO_2	% U_3O_8	% ThO_2	% U_3O_8
Polycrase Euxenite	5.0	7.30	4.65	6.95	6.60	6.45	3.45	7.82
Monazite	4.6	0.341	4.29	0.467	4.33	0.95	3.59	0.87
Thorianite A	64.8	25.2	68.0	26.0	70.2	25.0	67.4	25.2
Thorianite B	37.5	6.3	35.4	5.08	45.0	5.7	29.8	5.9
Thorite Orangite	59.9	7.8	63.6	8.1	61.2	1.78	48.9	2.98
Aeschynite Blomstrandine	5.88	4.2	6.60	3.86	9.66	3.06	5.73	4.27

* Analyses by USGS. All other analyses are by MIT.

The deviations of the radioassays from the chemical analyses in the preceding tables are much smaller than those obtained by the method of analysis based upon the self-absorption of beta radiation (MITG-208) but the final gamma-beta analyses are in no better agreement with the chemical analyses than are the preliminary results given in Table 3. It will be noted that the data in Table 3 show that the radiometric analyses tend to indicate ThO_2 contents which are too high and U_3O_8 contents which are too low, while the reverse is true of the preliminary data given in Table 4. Although the USGS has made no committment regarding the accuracy of the analyses it submitted, we believe that they are probably not in error by as much as ± 5 per cent. The uranium analyses also deviate from the true values but, in general, it is believed that such deviation should not exceed about ± 10 per cent. In view of these data it does not seem possible to conclude that inaccurate chemical analyses are largely responsible for the disagreement among the results obtained by the two methods. Some error may be attributed to the fact that the solutions of the simultaneous equations given in the Appendix sometimes involve relatively small differences between large numbers (on the average, any error in the measurement tends to be increased by a factor of about three when the per cent ThO_2 or U_3O_8 is computed). There is also the possibility that the radioactive constituents of many of the ores analyzed are not in equilibrium.

V. APPRAISAL OF THE GAMMA-BETA ACTIVITY METHOD

Radiometric discrimination between thorium and uranium by the gamma-beta method described in this report is much more accurate than by the self-absorption method described in Topical Report MITG-208. About half of the values obtained radiometrically were within 15 per cent of chemical values and practically all values were within a factor of 2. Although the radiometric method is apparently not an accurate analytical method, it should have some usefulness for appraising unknown samples where equipment is available.

The method does seem to show sufficient promise to warrant further investigation and some more careful consideration should be given to the problem of determining what sort of errors are likely to be inherent to it.

APPENDIX

A brief resume of the method used to calculate the per cent ThO_2 and U_3O_8 in the unknown mixtures is given below. The constants U_β , U_γ , T_β and T_γ , which may be regarded as the specific beta and gamma activities of uranium and thorium respectively, were determined with considerable care by measurements on materials whose composition was supposedly known precisely.

Uranium Constants: At first, two samples of pitchblende, containing negligible amounts of thorium, from the Eldorado mine were used to determine U_β and U_γ , with the following results:

Sample	U_β (cpm/g U_3O_8)	U_γ ($\times 10^{-13}$ amp/g U_3O_8)
NW-194 (1.32 per cent U_3O_8)	75,400	15.9
NW-208 (0.806 per cent U_3O_8)	75,000	15.7

It was then recalled that these Eldorado pitchblendes are not always in complete radioactive equilibrium, accordingly two samples of Congo ore were substituted, with the following results:

Sample	U'_β (cpm/g U_3O_8)	U'_γ ($\times 10^{-13}$ amp/g U_3O_8)
"G" Ore (1.81 per cent U_3O_8)	62,000	14.5
"C" Ore (3.49 per cent U_3O_8)	59,200	14.3

The second set of beta ray measurements was made with a different counter from that used for the first, hence the lower values for U'_β . Of course, the value for U_β used in calculating the assay of a sample was that obtained for the same counter on which the sample was run. There are, therefore, two sets of radioassays corresponding to the two counters and two values for the constants. However, most of the samples of a particular mineral used in both sets were not identical so that cross-comparison is not strictly possible.

Thorium Constants: An independent determination of T_β and T_γ was not possible because no thorium minerals containing negligible proportions of uranium are known. An error in the uranium constants would thus be carried over and magnified in the thorium constants. The material used for the determination of the thorium constants was a monazite sand, diluted with dunite, furnished by the National Bureau of Standards for use as a chemical and radiometric standard. It contained 1.138 per cent ThO_2 and 0.0448 per cent U_3O_8. Substitution of the values for U_γ , U_β and the fractions of ThO_2 and U_3O_8 in the following equations:

$$P_t T_\gamma + P_u U_\gamma = I = \text{gamma activity}$$

$$P_t T_\beta + P_u U_\beta = N = \text{beta activity}$$

gave $T_\beta = 16{,}000$, $T_\gamma = 7.0$ for the preliminary set and $T'_\beta = 12{,}850$, $T'_\gamma = 7.05$ for the final set (units for U_β , U_γ , T_β , T_γ are given on this page and are the same throughout). It is seen that the ratios $T_\beta : U_\beta$ and $T'_\beta : U'_\beta$ are the same to within 0.5 per cent.

Analysis of Unknown Mixtures:

Having determined the specific beta and gamma activities of uranium and thorium one can analyze an unknown mixture of both elements after making separate beta and gamma measurements. The standard solutions for the simultaneous equations give

$$\%U_3O_8 = \frac{N_x - 2286I_x}{39,000} \times 100$$

$$\%ThO_2 = \frac{4740I_x - N_x}{17,200} \times 100$$

for the preliminary set and

$$\%U_3O_8 = \frac{N'_x - 1825I'_x}{34,300} \times 100$$

$$\%ThO_2 = \frac{4210\ I'_x - N'_x}{16,850} \times 100$$

for the final set. I_x, N_x, I'_x and N'_x, again are the measured gamma and beta activities for the first and second sets of unknown mixtures, respectively.

The experimental data in the preliminary set are summarized in Table A1.

Table A1. Experimental Data for the Preliminary Analysis of Mixtures

Sample	Ionization Current Per Gram	Beta Counting Rate
Standards:		
N.B.S. Monazite	0.087×10^{-13} amp	215 cpm
NW-194 Eldorado pitchblende	0.21	995
NW-208 Eldorado pitchblende	0.13	605
Unknowns:		
Polycrase Euxenite (1/5)	0.29	1178
Thorite Orangite (1/60)	0.076	185
Thorianite B (1/4)	1.03	2814
Thorianite B (1/15)	0.27	764
Thorianite A (1/6)	1.48	4997
Thorianite A (1/15)	0.51	1377
Monazite (1/3)	0.148	382
Monazite	0.41	1215
Aeschynite Blomstrandine (1/6)	0.19	1377

Figures in parentheses show the extent to which the minerals were diluted with silica before their activities were measured.

The results obtained from the second suite of samples, and with a different Geiger tube, are given in Table A2.

Table A2. Experimental Data for the Final Analysis of Mixtures

Sample	Ionization Current Per Gram	Beta Counting Rate
Standards:		
N.B.S. Monazite	0.0867×10^{-13} amp	173 cpm
"C" Ore	0.502	2065
"G" Ore	0.263	1121
Unknowns:		
Polycrase Euxenite	1.38	5230
Thorite Orangite (1/30)	0.13	271
Thorianite B (1/15)	0.197	495
Thorianite A (1/20)	0.42	1198
Monazite	0.38	995
Aeschynite Blomstrandine	1.03	3365
Non Mag Fraction Black Sand A-5	0.04	92.7
Non Mag Fraction Black Sand A-21	0.0273	74.9
Non Mag Fraction Black Sand A-23	0.367	912
Non Mag Fraction Black Sand A-25	0.022	55.5
Non Mag Fraction Black Sand A-26	0.19	471
Euxenite	2.6	10,600

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